

PATENT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN THE APPLICATION OF:

DONALD BERNARD BIVENS ET. AL.

CASE NO.: FL1065 US NA

SERIAL NO.: 09/528,964

GROUP ART UNIT: 1751

FILED: MARCH 20, 2002

EXAMINER: HARDEE, JOHN R.

FOR: COMPOSITIONS OF DIFLUOROMETHANE,  
PENTAFLUOROETHANE, 1,1,1,2-TETRAFLUOROETHANE  
AND HYDROCARBONS

---

**DECLARATION OF BARBARA H. MINOR UNDER 37 C.F.R. 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, BARBARA H. MINOR, declares as follows:

I am a citizen of the United States of America.

I reside at 233 Greenhaven Drive, Elkton, Maryland 21921.

I received a Bachelors of Science degree in Chemical Engineering  
from the Bucknell University in 1981.

I am an employee of E. I. du Pont de Nemours and Company (hereinafter  
"DuPont"), 1007 Market Street, Wilmington, Delaware 19898, USA :

Presently, I am employed as a Senior Research Associate for the  
Fluoroproducts business of DuPont, which is located in Wilmington,  
Delaware, USA.

I joined DuPont in 1981. Since that time, I have held various  
assignments in research, manufacturing support and technical service. I have  
also held technical positions in specialty chemicals and fluorochemicals. In

1989, I commenced research to identify new alternatives to chlorofluorocarbons (hereinafter “CFC”) refrigerants. Over the past thirteen (13) years, I have obtained approximately fifty (50) patents, published numerous refrigerant-related materials and made many presentations in the field of CFC alternative refrigerants.

During my tenure with DuPont Fluoroproducts, I have also held the following positions:

- Chairman of ASHRAE (American Society of Heating, Refrigeration, and Air Conditioning Engineers) TG10.MOC Immiscible Oil-Refrigerants subcommittee
- Secretary of ASHRAE TC3.1 Refrigerants thermophysical properties subcommittee
- Member of ARI 21<sup>st</sup> Century Working Fluids subcommittee
- Member of ARI ISO Refrigerants Task Group

I, am a named as one of the inventors in US Patent Application No. 09/528,964, entitled “Compositions of Difluoromethane, Pentafluoroethane, 1,1,1,2-Tetrafluoroethane and Hydrocarbons”, and am familiar with its contents.

More particularly, I am providing the following statements and data to demonstrate the novelty and non-obviousness of the inventive subject matter of US Patent Application No. 09/528,964.

This affidavit is prepared for the purpose of presenting technical information addressing the novelty of the present patent application of JP 9-25480 and non-obviousness in view of JP 9-25480 and WO 96/03473 A1.

Documents provided herein, Attachment 1 and Attachment 2 were prepared according to the method described therein.

The explanation on the technical meaning was prepared to the best of my knowledge and experience in the field relevant to the subject matter.

## **ATTACHMENT 1**

### **Composition Diagram Comparison**

#### **Method to Prepare Composition Diagram:**

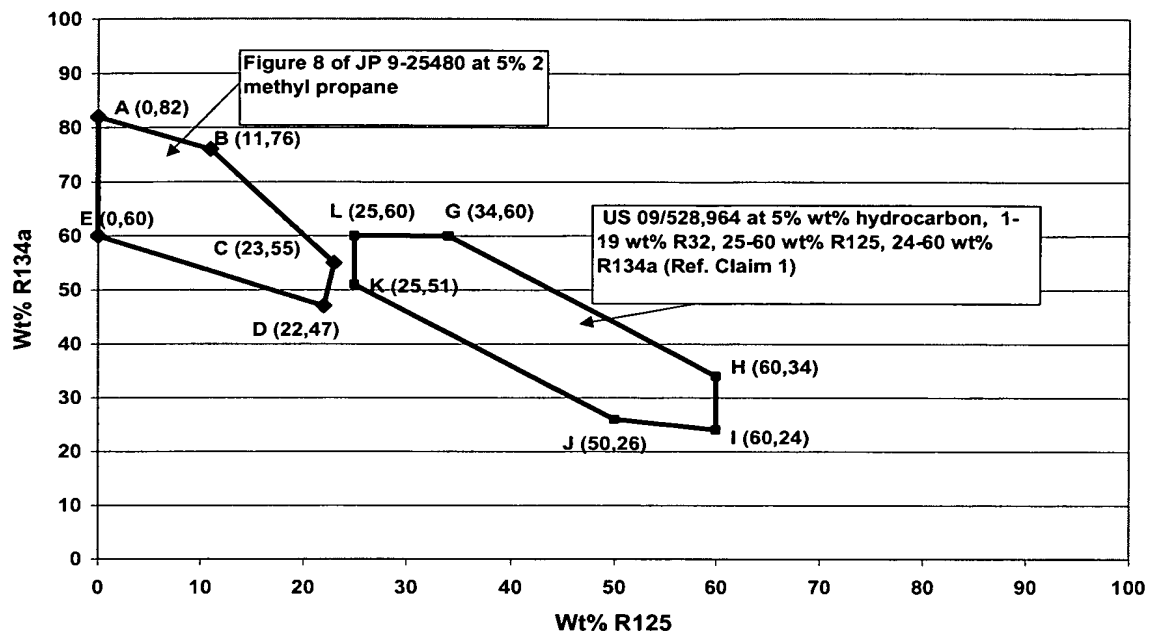
JP 9-25480 exemplifies composition of HFC-32, HFC-125, HFC-134a and 2-methyl propane in Figures 8, 14, 20, 26 and 32 of the specification. Figure 8 shows compositions containing 5 wt% 2 methyl propane, Figure 14 at 10 wt%, Figure 20 and 15 wt%, Figure 26 at 20 wt% and Figure 32 at 25 wt% respectively. Since hydrocarbons are known to be highly flammable, Figure 8 represents the least flammable compositions in the specification. Figure 8 of JP 9-25480 was redrawn as Figure 1 below with exemplified compositions enclosed by points A to E. The x-axis depicts wt% R125 and the y-axis depicts wt% R134a. Assuming all compositions contain 5 wt% 2-methyl propane, which represents the least flammable composition disclosed in the specification, the amount of R32 in the composition is the remainder.

The compositions in Claim 1 of US Patent Application No. 09/528,964 contain about 1 to about 19 wt% HFC-32, from about 25 to about 60 wt% HFC-125, from about 24 to about 60 wt% R134a and from about 0.5 to about 5 wt% of a hydrocarbon selected from the group consisting of: n-butane, isobutane (e.g. 2-methyl propane); n-butane and 2-methyl butane; n-butane and n-pentane; isobutane and 2-methyl butane; and isobutane and n-pentane. Compositions of Claim 1 of US No. 09/528,964 were drawn in Figure 1 enclosed by points G to L wherein the hydrocarbon composition was held constant at 5 wt% to be consistent with JP 9-25480.

#### **Technical Explanation of Figure 1 – Composition Diagram**

JP 9-25480 exemplifies compositions with the lowest amount of hydrocarbon at 5 wt% in Figure 8. Claim 1 of US No. 09/528,964 recites a hydrocarbon range of 0.5 to 5 wt%. Therefore, the only exemplified compositions in the reference where hydrocarbon amounts are equivalent to the present invention is at 5 wt% hydrocarbon, which are compositions shown in Figure 1 below. The compositions enclosed by points A to E from JP 9-25480 and points G to L for US No. 09/528,964 do not overlap at any point.

Figure 1 - Composition Comparison



## ATTACHMENT 2

### TEH Comparison to Evaluate Refrigerant Flammability

#### **Experimental Method:**

A method to estimate flammability of a refrigerant composition containing R32, R125 and hydrocarbons called TEH (Total Equivalent Hydrocarbon) was developed and described in US 09/528,964 as follows: Based on standard flammability test method ASTM 681 at 100°C, the following flammability limits have been determined:

<b>Composition</b>	<b>Flammability Limit (Wt%)</b>
HFC-125/HFC-32	57 % HFC-32
HFC-134a/HFC-32	33 % HFC-32
HFC-125/n-butane	6 % n-butane
HFC-134a/n-butane	3 % n-butane

The data show compositions with a higher amount of HFC-125 can tolerate more hydrocarbon and still be nonflammable. Also, HFC-32 is about 10 times less flammable than hydrocarbons. To give an indication of mixture flammability, the following formula gives an approximation of the “total equivalent hydrocarbon” (TEH) present in mixtures that contain both HFC-32 and hydrocarbons:  $TEH = HC + R32/10$ , where TEH = Total Equivalent Hydrocarbon in weight percent, HC = weight percent hydrocarbon in a mixture, and R32 = weight percent HFC-32 in a mixture. For the compositions of the present invention, it is useful to relate the amount of HFC-125 in the mixture to flammability because HFC-125 has some degree of flame suppression. Table 1 indicates the flammability limit of a mixture containing both HFC-32 and hydrocarbons based on HFC-125 composition and TEH.

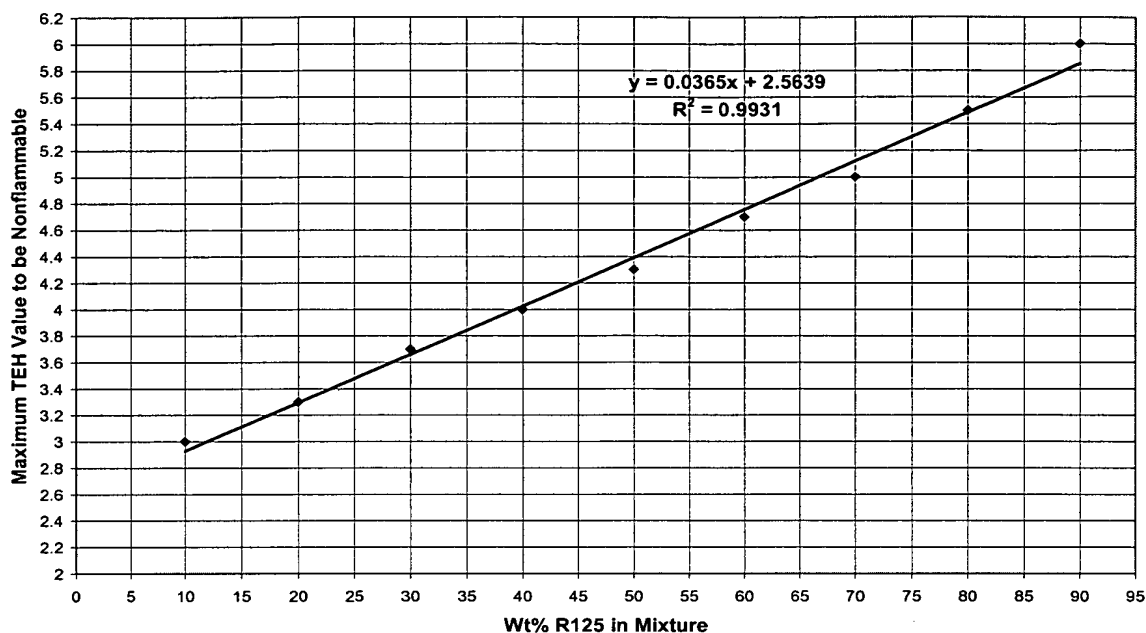
**TABLE 1**

Weight Percent HFC-125 in HFC-32/HFC-125/HFC- 134a/HC Mixture	Maximum Weight Percent TEH To Be Nonflammable
10	3.0
20	3.3
30	3.7
40	4.0
50	4.3
60	4.7
70	5.0
80	5.5
90	6.0

**TEH Comparison for US 09/528,964, WO 9603473 and JP 9-25480:**

To calculate and compare TEH values for compositions exemplified in the above references, a linear regression was developed for TEH based on values in Table 1. The regression is shown in Figure 2 where the maximum value for TEH to be nonflammable is approximately equal to  $(\text{Wt\% R125}) * 0.0365 + 2.5639$ .

Figure 2 - Wt% R125 Versus TEH (Table 1 of US No. 09/528,964 )



TEH values are already shown in Tables 2 and 3 of US 09/528,964 and demonstrate the exemplified compositions are non-flammable as originally formulated and even after a 97% vapor leak. These data satisfy one of the objectives of the invention and meet industry requirements for non-flammable refrigerant formulations. WO 96/03473 exemplifies compositions containing R32, R125 and hydrocarbons, optionally with R134a in Examples 1-6 and 13-18. TEH values were calculated and compared with the linear regression value for the maximum TEH allowed for non-flammability (based on R125 composition). Results are shown in Table 2. All compositions exemplified in WO 9603473 Examples 1-6 and 13-18 are flammable by TEH analysis which differentiate them from US 09/528,964 as less preferred refrigerant formulations.

**Table 2 – TEH Calculations for Compositions Exemplified in WO 96/03473**

Example No.	Wt% R32	Wt% R125	Wt% R134a	Wt% Hydrocarbon	TEH Value of Example	Max TEH to be non-flammable based on wt% R125	Flammable?
1	22.5	24.5	51.0	2.0	4.3	3.5	Yes
2	22.1	24.0	49.9	4.0	6.2	3.4	Yes
3	21.6	23.5	48.9	6.0	8.2	3.4	Yes
4	49.0	49.0	0.0	2.0	6.9	4.4	Yes
5	48.0	48.0	0.0	4.0	8.8	4.3	Yes
6	47.0	47.0	0.0	6.0	10.7	4.3	Yes
13	22.5	24.5	51.0	2.0	4.3	3.5	Yes
14	22.1	24.0	49.9	4.0	6.2	3.4	Yes
15	21.6	23.5	48.9	6.0	8.2	3.4	Yes
16	49.0	49.0	0.0	2.0	6.9	4.4	Yes
17	48.0	48.0	0.0	4.0	8.8	4.3	Yes
18	47.0	47.0	0.0	6.0	10.7	4.3	Yes

JP 9-25480 exemplifies compositions containing R32, R125, R134a and 2-methyl propane in Figures 8, 14, 20, 26 and 32. TEH values were calculated at composition points shown in each figure and compared with the linear regression value for the maximum TEH allowed for non-flammability based on R125 composition. Results are shown in Table 3. All compositions exemplified in JP 9-25480 Figures 8, 14, 20, 26 and 32 are flammable by TEH analysis which differentiates them from US 09/528,964 as less preferred refrigerant formulations.

**Table 3 - TEH Calculation for Compositions Exemplified in JP 9-25480**

Point	wt% R125	wt % R134A	wt% R32	wt% 2-methyl propane	TEH Value at each point in figure	Max TEH to be non-flammable based on linear regression	Flammable?
Figure 8							
A	0	82	13	5	6.3	2.6	Yes
B	11	76	8	5	5.8	3.0	Yes
C	23	55	17	5	6.7	3.4	Yes
D	22	47	26	5	7.6	3.4	Yes
E	0	60	35	5	8.5	2.6	Yes
F	15	65	15	5	6.5	3.1	Yes
Figure 14							



A	0	81	9	10	10.9	2.6	Yes
B	19	55	16	10	11.6	3.3	Yes
C	19	48	23	10	12.3	3.3	Yes
D	13	52	25	10	12.5	3.0	Yes
E	0	60	30	10	13.0	2.6	Yes
F	0	72	18	10	11.8	2.6	Yes
P	9	60	21	10	12.1	2.9	Yes
Figure 20							
A	0	79	6	15	15.6	2.6	Yes
B	27	39	19	15	16.9	3.6	Yes
C	7	52	26	15	17.6	2.8	Yes
D	0	56	29	15	17.9	2.6	Yes
E	0	50	35	15	18.5	2.6	Yes
Figure 26							
A	0	73	7	20	20.7	2.6	Yes
B	20	50	10	20	21.0	3.3	Yes
C	20	37	23	20	22.3	3.3	Yes
D	0	52	28	20	22.8	2.6	Yes
E	0	63	17	20	21.7	2.6	Yes
Figure 32							
A	0	66	9	25	25.9	2.6	Yes
B	12	56	7	25	25.7	3.0	Yes
C	19	43	13	25	26.3	3.3	Yes
D	19	38	18	25	26.8	3.3	Yes
E	7	38	30	25	28.0	2.8	Yes
F	0	44	31	25	28.1	2.6	Yes

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Wherefore, this affidavit is hereby executed on this 1st day of March 2004.

  
Barbara Haviland Minor



## Certificate of Mailing under 37 CFR 1.8

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to:

Commissioner for Patents; Mail Stop: AF; P.O. Box 1450  
Alexandria, VA 22313-1450

on March 1, 2004  
Date

Barbara A. Ferzetti  
Signature

BARBARA A. FERZETTI  
Type or printed name of person signing Certificate

Note: Each paper must have its own certificate of mailing, or this certificate must identify each submitted paper.

09/528,964  
FL-1065 US NA

Response  
Declaration of Barbara H. Minor under 37 CFR 1.132  
Post Card Receipt